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(71) Applicant:

00002174

Sekisui Kagaku Kogyo K. K. [Sekisui
Chemical Co., Ltd.]

Nishitemma 2 Chome 4 Ban 4 Go
Kita Ku, Osaka Shi, Osaka Fu

(72) Inventor:

Nobuhiro Goto

Sekisui Kagaku Kogyo K. K.
Kamitoba, Kami Choshi Cho 2-2
Minami Ku, Kyoto Shi

(72) Inventor:

Masashi Nakadani
Sekisui Kagaku Kogyo K. K.
Kamitoba, Kami Choshi Cho 2-2
Minami Ku, Kyoto Shi

(72) Inventor:

Takeharu Morita
Sekisui Kagaku Kogyo K. K.
Shimamoto Cho, Hyakusan 2-1
Mishima Gun, Osaka Fu

(72) Inventor:

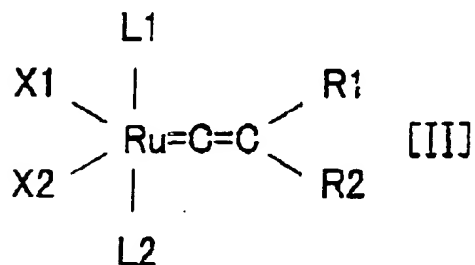
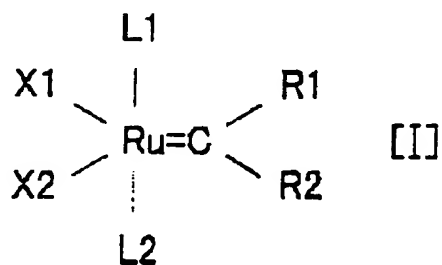
Masaya Kobayashi
Sekisui Kagaku Kogyo K. K.
Kamitoba, Kami Choshi Cho 2-2
Minami Ku, Kyoto Shi

(54) [Title Of The Invention] Adhesive For Olefin Resins

(57) [Abstract]

[Problem] To be able to adhere sets of olefin resins firmly and in a short time.

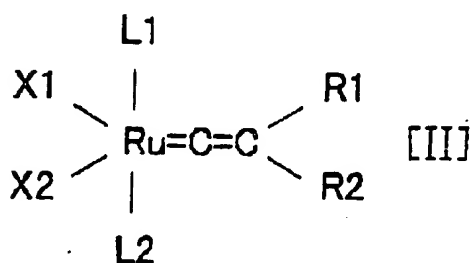
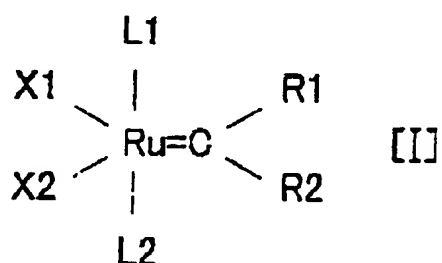
[Means Of Resolving The Problem] To construct a solventless reactive adhesive whose indispensable component is a norbornene monomer that is an olefin monomer and either a ruthenium-carbene complex as in general formula [I] below or a ruthenium vinylidene complex as in general formula [II] below.



[Scope Of Patent Claim]

[Claim Paragraph 1] Adhesive for olefin resins characterized in that it is constructed of a norbornene monomer and either a ruthenium-carbene complex as in general formula [I] below or a ruthenium vinylidene complex as in general formula [II] below as an indispensable ingredient.

[Chemical 1]



[Claim Paragraph 2] Adhesive for olefin resins described in Claim Paragraph 1 characterized in that the norbornene monomer is a norbornene monomer of 3 rings or more.

[Claim Paragraph 3] Adhesive for olefin resins described in Claim Paragraphs 1 and 2 characterized in that the double bond contains 2 or less unsaturated single ring cycloolefins.

[Claim Paragraph 4] Adhesive for olefin resins described in Claim Paragraph 3 characterized in that the unsaturated single ring cycloolefins are dienes.

[Detailed Explanation Of The Invention]

[0001]

[Industrial Field To Which The Invention Belongs] The present invention relates to an adhesive for olefin resins.

[0002]

[Prior Technology] Because olefins such as polypropylene (PP) and polyethylene (PE) have little polarity and do not take functional groups on their surfaces, they are known as the most difficult plastics to adhere. Adhesives to adhere sets of such olefin resins that have been offered are solvent type adhesives and hot melt adhesives.

[0003] Also, a method of adhering sets of olefin adhesives that has been offered is the method by instant adhesion after treating with primer, as disclosed in Japanese Patent Disclosure Sho 62-18485.

[0004]

[Problems To Be Resolved By The Invention] In addition to the fact that solvent type adhesives are harmful to humans because of their solvents, it takes time for the solvent to volatize, and the adhesive strength it self is not sufficient.

[0005] Hot melt adhesives require apparatus and work to melt the adhesive. These adhesives also do not give sufficient adhesive strength. Further, although using hot melt adhesives with preheating of the olefin resins increases adhesive strength, the olefin resins end up deformed and this leads to a new problem.

[0006] Following the adhesion method disclosed in Japanese Patent Disclosure Sho 62-18485 not only involves the work of primer treatment by also fails to achieve sufficient adhesive strength.

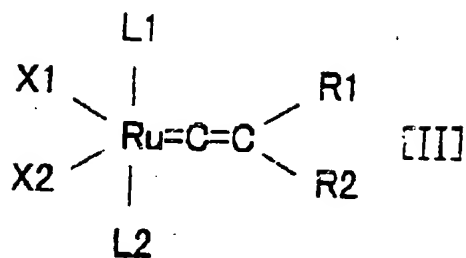
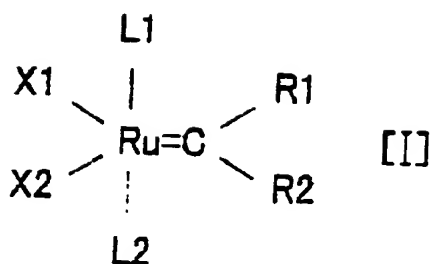
[0007] The present invention is one that was made with these facts in view, and its object lies in offering a superior adhesive for olefin resins that can show sufficient adhesive strength on olefins in short time periods in a solventless environment.

[0008]

[Means For Resolving The Problems] The adhesive for olefin resins of the present invention is characterized by a construction having a norbornene monomer and either a ruthenium-carbene complex of general formula [I] below or a ruthenium vinylidene complex of general formula [II] below as an indispensable component.

[0009]

[Chemical 2]



[0010] The norbornene monomers used in the adhesive for olefin resins of the present invention can be cyclics like bicyclics such as norbornene or norbornene diene, tircyclics such as dicyclopentadiene or dihydrocyclopentadiene, tetracyclics such as tetracyclodecene, ethylidene tetracyclodecene and phenyltetracyclodecene, pentacyclics such as tricyclopentadiene and heptacyclics such as tetracyclopentadiene, and of course their alkyl substitutions (for example, such as methyl, ethyl, propyl and butyl substitutions), alkylidene substitutions (for example, such as ethylidene substitutions) and aryl substitutions (for example, phenyl and tolyl substitutions), with consideration for their adhesiveness to various types of olefin resins comprising the adhered members, and

it is also possible to use derivatives having polar groups such as epoxy groups, methacrylic groups, hydroxide groups, amino groups, carboxyl groups, cyano groups, halogen groups, ether groups and groups containing ester bonds. Although norbornene monomers having such polar groups did not have polymerization that advanced well with prior catalysts, it is possible to complete the polymerization by using as the polymerization catalyst the ruthenium-carbene complex of general formula [I] or the ruthenium vinylidene complex of general formula [II] of the present invention, and it is possible to form good adhesive interfaces with the olefin resins that are the adhered members. These norbornene monomers can be used singly or used in mixtures of 2 or more.

[0011] By blending the ruthenium-carbene complex of general formula [I] or the ruthenium vinylidene complex of general formula [II] into such norbornene monomers, it becomes possible to make norbornene polymer by advancing the ring opening polymerization that is called metathesis polymerization.

[0012] Among the said norbornene monomers, using norbornene monomers of 3 rings or more as the adhesive component will give norbornene polymers that become crosslinked polymers which further increase adhesive strength. It is preferred that this monomer be dicyclopentadiene from the standpoints of cost, reactivity, properties and easy obtainability.

[0013] Among the adhesives for olefin resins of the present invention, using unsaturated single ring cycloolefins where the

double bonds are 2 or less mixed with the norbornene monomer makes it possible to control the metathesis polymerization reaction speed and is beneficial in controlling pot life.

[0014] Unsaturated single ring cycloolefins that can be mentioned are those with 1 double bond such as cyclobutene, cyclopentene, cyclooctene and cyclodecene and those with 2 double bonds such as cyclopentadiene and cyclooctadiene, among which cyclooctadiene is particularly preferred. The amount contained should be in the range of 0.1 to 50 mol% of the total polymerizable monomer. At less than 0.1 mol% there are cases when pot life cannot be regulated, and when 50 mol% is exceeded there are cases when polymerization speed becomes too slow and polymerization ends up incomplete.

[0015] Viscosity regulation agents may be blended into the adhesive for olefin resins of the present invention, including such as dipolyisoprene and polybutadiene having olefinic unsaturated groups, and such viscosity regulators can be put into the polymer at time of metathesis polymerization. The amount of viscosity regulator contained is preferably in the range of 0.1 to 50 wt% of the total polymerizable monomer, and more preferably in the range of 0.1 to 20 wt%.

[0016] Next, the ruthenium-carbene complex of general formula [I] will be explained in further detail.

[0017] In the formula, R1 and R2 can be substituted mutually independently as required by hydrogen, C2 to C20-alkenyl, C1 to C20-alkyl, aryl, C1 to C20-carboxylate, C1 to C20-alkoxy, C2 to C20-

alkenyloxy, C2 to C20-alkoxycarbonyl, C1 to C20-alkylthio (these may be substituted as required by C1 to C5-alkyl, halogens and C1 to C5-alkoxy, or they may be substituted as required by phenyl substituted by C1 to C5-alkyl, halogens and C1 to C5-alkoxy). X1 and X2 in the formula are mutually independently any anionic ligand, and L1 and L2 are mutually independently any neutral electron donor. Further, two or three of X1, X2, L1 and L2 may together form a multidentate chelate ligand.

[0018] Preferred ruthenium-carbene complexes shown by Formula [I] are ruthenium-carbene complexes where R1 and R2 in the formula are mutually independently hydrogen, C2 to C5-alkenyl, C1 to C5-alkyl, phenyl, C1 to C5-carboxylate, C1 to C5-alkoxy, phenoxy, C2 to C5-alkoxycarbonyl (these may be substituted as required by C1 to C5-alkyl, halogens and C1 to C5-alkoxy, or they may also be substituted as required by phenyl substituted by C1 to C5-alkyl, halogens and C1 to C5-alkoxy), where X1 and X2 are anionic ligands that may mutually independently be substituted as required by Cl, Br, C1 to C5-carboxylate, C1 to C5-alkoxy, phenoxy and C1 to C5-alkylthio (these may be substituted as required by C1 to C5-alkyl, halogens and C1 to C5-alkoxy, or they may be substituted as required by phenyl substituted by C1 to C5 alkyl, halogens and C1 to C5-alkoxy), and where L1 and L2 are neutral electrons that may mutually independently be substituted as required by aryl or C1 to C10-alkylphosphine (these may be substituted as required by C1 to C5-alkyl, halogens and C1 to C5-alkoxy, or may be substituted as

required by phenyl substituted by C1 to C5-alkyl, halogens and C1 to C5-alkoxy.

[0019] Preferred ruthenium-carbene complexes of general formula [I] are ruthenium-carbene complexes where R1 and R2 are mutually independently hydrogen, methyl, ethyl and phenyl, or are vinyl substituted as required by methyl, ethyl or phenyl, where X1 and X2 are mutually independently Cl or Br, and where L1 and L2 are mutually independently trimethylphosphine, triethylphosphine, triphenylphosphine or tricyclohexylphosphine.

[0020] Next the ruthenium vinylidene complex of general formula [II] will be explained in further detail.

[0021] In the formula, R1 and R2 are mutually independently the ferrocene derivatives hydrogen, C2 to C20-alkenyl, C1 to C20-alkyl, aryl, C1 to C20-carboxylate, C1 to C20-alkoxy, C2 to C20-alkenyloxy, C2 to C20-alkoxycarbonyl, C1 to C20-alkylthio (these may be substituted as required by C1 to C5-alkyl, halogens and C1 to C5-alkoxy, or they may be substituted as required by C1 to C5-alkyl, halogens and C1 to C5-alkoxy). X1 and X2 in the formula are mutually independently any anionic ligand, and L1 and L2 are mutually independently any neutral electron donor. Further, two or three of X1, X2, L1 and L2 may together form a multidentate chelate ligand.

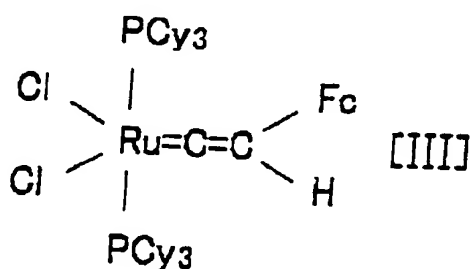
[0022] The more preferred ruthenium vinylidene complexes of general formula [II] are ruthenium vinylidene complexes where R1 and R2 in the formula are mutually independently hydrogen, methyl,

ethyl, phenyl and ferrocenyl, or substitutions as required with methyl, ethyl, phenyl or ferrocenyl, where X1 and X2 are mutually independently Cl or Br, and where L1 and L2 are mutually independently trimethyl phosphine, triethyl phosphine, triphenyl phosphine or tricyclohexyl phosphine.

[0023] Further, the complex of the following structural formula [III] can be offered as a specific example of the ruthenium vinylidene complex.

[0024]

[Chemical 3]



Fc: フェロセニル基

Fc: ferrocenyl group

[0025] Prior polymerization catalysts for metathesis polymerization of norbornene monomers are combinations of catalysts and catalytic activators, the catalysts being such as halogen compounds, oxyhalogen compounds, oxides and organic ammonium salts of such as tungsten, molybdenum, rhenium and tantalum, and the activators being such as alkyl aluminum halide, alkoxyalkyl aluminum halide, aryloxyalkyl aluminum halide and organic tin compounds. In the

case of these catalyst systems, the norbornene monomer polymerization reaction was hindered by the presence of oxygen and moisture, and unless done in a completely inert gas atmosphere there were problems in advancing the polymerization, so that it became impossible to apply them as adhesives.

[0026] The ruthenium-carbene complex of general formula [I] and the ruthenium vinylidene complex of general formula [II] are stable under moisture, and by blending them with norbornene monomer it becomes possible use them as adhesives for olefin resins with no problems, even if the work is done outdoors or in the rain.

[0027] In regard to the ruthenium-carbene complex of general formula [I] or the ruthenium vinylidene complex of general formula [II], they may be mixed directly with the norbornene monomer; or they may be dispersed as liquid in the norbornene monomer. As an example, they can be used diluted in a solvent. Examples of solvents are such as toluene, benzene, tetrahydrofuran (THF), dichloromethane or liquid hydrocarbons such as process oil. It is not preferred to dissolve the ruthenium-carbene complex of general formula [I] and the ruthenium vinylidene complex of general formula [II] in acid or solvents where active moisture is present, since there will be loss of complex stability. Although the ruthenium vinylidene complex of general formula [II] has higher stability in air than the ruthenium-carbene complex of general formula [I], they can be said to be about the same.

[0028] The mixture ratio of the ruthenium-carbene complex of general formula {I} and the ruthenium vinylidene complex of general formula {II} in the norbornene monomer or monomer mixture of the norbornene monomer with unsaturated monocyclic cycloolefin monomer (hereafter, called polymerizable monomer) is preferably in the range of 1/5 to 1/500000 mol. When the mixture ratio exceeds [sic] 1/5 mol the reaction becomes too fast and there is not enough time for the adhesion work, and when the mixture ratio is less than [sic] 1/500000 there will be insufficient hardening of the polymerizable monomer. A more preferred mixture ratio is in the range of from 1/30 to 1/200000. This range will display sufficient adhesive strength and good reactivity.

[0029] Methods of using the adhesive for olefin resins of the present invention include the method of painting on the substrate with such as a hair brush and the method of blowing on it with spray.

[0030] When a spray is used, the preferred type is one that sprays in air the polymerizing composition with the ruthenium-carbene complex of general formula [I] or the ruthenium vinylidene complex of general formula [II] diluted in solvent, being preferred from the standpoints of workability and quality, and a market spray gun can be utilized.

[0031] The olefin resins that can be adhered with the adhesive of the present invention include norbornene polymers such as polydicyclopentadiene and polynorbornene and their derivatives, their hydrogen additions of course, low density polyethylene

(LDPE), linear low density polyethylene (LLDPE), high density polyethylene (HDPE), polypropylene (PP), polybutene and polyisoprene. It can also be used on ethylene-vinyl acetate copolymer, ethylene-ethylacrylate copolymer, ionomers, and terpolymers such as ethylene-propylene-dienes.

[0032] It is preferred that the paint thickness of such adhesives for olefin resins be 1 mm or more. Having the paint thickness 1 mm or more makes it possible to utilize the polymerization exothermicity during polymerization of the norbornene monomer and to fuse the olefin resin to form a stronger adhesion interface. Also, performing heat treatment of the adhesion surface at 40 to 90°C during adhesion or after adhesion makes it possible to advance the hardening reaction and further increase adhesion strength. The means for doing this may be to use a heating furnace or to heat with such as a drier. However, since norbornene polymers such as polydicyclopentadiene and polynorbornene as well as their derivatives and hydrogen addition products are the same norbornene materials, it is possible to realize good adhesive strength without relying on paint thickness.

[0033] Further, various types of additives such as elastomers may be blended into the adhesive for polyolefin resins of the present invention, to serve as thixotropy imparters, antioxidants, fillers, colorants, high molecular reformers, flame retardants, foamers, thixotropy imparters [sic], antistatic agents, molecular weight regulators, pigments, dyes and shock resistance imparters.

[0034] The fillers may be such as calcium carbonate, aluminum hydroxide, magnesium carbonate, sodium hydrogen carbonate, clay, talc, mica, kaolin, fly ash, montmorillonite, glass balloon, silica balloon and thermal expanding vinylidene chloride particles.

[0035] The elastomers may be such as natural rubber, polybutadiene, polyisoprene, styrene-butadiene-styrene block copolymer, styrene-isoprene-styrene block copolymer, EPDM, ethylene-vinyl acetate copolymer and their hydrides.

<Action> By following the adhesive of the present invention, it is possible to adhere olefin resins quickly and with superior adhesive strength. Further, since the norbornene monomer that is an indispensable ingredient is itself an olefin monomer, it is possible to form with good affinity a good adhesion interface with olefin resins.

[0036]

[Examples] <Example 1>

Polymerizable monomer: 90 mol% dicyclopentadiene (90.8 wt%), 10 mol% 5-ethylidene-2-norbornene (9.2 wt%),

Ruthenium-carbene complex: bis(tricyclohexylphosphine)benzylidene ruthenium dichloride.

A 2 mm thick polypropylene sheet was prepared, and after surface degreasing, was cut into two pieces 25 mm X 100 mm. The end of one of these cut pieces (area: 12.5 mm X 25 mm) was painted with an adhesive composition blended so that the mol ratio of the bis(tricyclohexylphosphine)benzylidene ruthenium dichloride was 1/10000 in respect to the polymerizable monomer, and then the

other piece had its end (area: 12.5 X 25 mm) laid over and pasted on, and the product was given heat treatment at 80°C for 5 minutes under application of 490 kPa pressure to make a test piece. The adhesion work time including heat treatment was 7 minutes.

[0037] <Example 2>

Polymerizable monomer: 90 mol% dicyclopentadiene (90.8 wt%), 10 mol% 5-ethylidene-2-norbornene (9.2 wt%),

Ruthenium vinylidene complex: complex of the aforesaid formula III. A 2 mm thick polypropylene sheet was prepared, and after surface degreasing, was cut into two pieces 25 mm X 100 mm. The end of one of these cut pieces (area: 12.5 mm X 25 mm) was painted with an adhesive composition blended so that the mol ratio of the ruthenium vinylidene complex was 1/5000 in respect to the polymerizable monomer, and then the other piece had its end (area: 12.5 X 25 mm) laid over and pasted on, and the product was given heat treatment at 80°C for 15 minutes under application of 490 kPa pressure to make a test piece. The adhesion work time including heat treatment was 17 minutes.

[0038] <Comparative Example 1> A 2 mm thick polypropylene sheet was prepared, and after surface degreasing, two 25 mm X100 mm cut pieces were made, the ends of the two cut pieces (areas: 12.5 mm X 25 mm) were dipped in a primer comprising a 5% acetone solution of 6-amino-2-picoline and then withdrawn, and were let stand for a while to volatize the acetone. Then, ethylcyanoacrylate was painted onto the primer treated surface (area: 12.5 mm X 25 mm) of one cut

piece, after which the primer treated surface (area: 12.5 mm X 25 mm) of the other cut piece was laid over and pasted, and the combined pieces were let stand for 5 hours under application of 490 kPa pressure to make a test piece. The adhesion work time extended to more than 5 hours.

[0039] <Comparative Example 2> A 2 mm thick polypropylene sheet was prepared, and after surface degreasing, two 25 mm X100 mm cut pieces were made, the ends of the two cut pieces (areas: 12.5 mm X 25 mm) were dipped in a primer comprising a 5% acetone solution of 4-dimethylaminopyridine and then withdrawn, and were let stand for a while to volatize the acetone. Then, ethylcyanoacrylate was painted onto the primer treated surface (area: 12.5 mm X 25 mm) of one cut piece, after which the primer treated surface (area: 12.5 mm X 25 mm) of the other cut piece was laid over and pasted, and the combined pieces were let stand for 5 hours under application of 490 kPa pressure to make a test piece. The adhesion work time extended to more than 5 hours.

[0040] <Example 3>

Polymerizable monomer: 90 mol% dicyclopentadiene (90.8 wt%), 10 mol% 5-ethylidene-2-norbornene (9.2 wt%),

Ruthenium-carbene complex:

bis(tricyclohexylphosphine)benzylidene ruthenium dichloride.

A 2 mm thick high density polyethylene sheet was prepared, and after surface degreasing, was cut into two pieces 25 mm X 100 mm.

The end of one of these cut pieces (area: 12.5 mm X 25 mm) was

painted with an adhesive composition blended so that the mol ratio of the bis(tricyclohexylphosphine)benzylidene ruthenium dichloride was 1/10000 in respect to the polymerizable monomer, and then the other piece had its end (area: 12.5 X 25 mm) laid over and pasted on, and the product was given heat treatment at 80°C for 5 minutes under application of 490 kPa pressure to make a test piece. The adhesion work time including heat treatment was 7 minutes.

[0041] <Example 4>

Polymerizable monomer: 90 mol% dicyclopentadiene (90.8 wt%), 10 mol% 5-ethylidene-2-norbornene (9.2 wt%),

Ruthenium vinylidene complex: complex of the aforesaid formula III.

A 2 mm thick polyethylene sheet was prepared, and after surface degreasing, was cut into two pieces 25 mm X 100 mm. The end of one of these cut pieces (area: 12.5 mm X 25 mm) was painted with an adhesive composition blended so that the mol ratio of the ruthenium vinylidene complex was 1/5000 in respect to the polymerizable monomer, and then the other piece had its end (area: 12.5 X 25 mm) laid over and pasted on, and the product was given heat treatment at 80°C for 15 minutes under application of 490 kPa pressure to make a test piece. The adhesion work time including heat treatment was 17 minutes.

[0042] <Example 5>

Polymerizable monomer: 90 mol% dicyclopentadiene (91.7 wt%), 10 mol% cyclooctadiene (8.3 wt%),

Ruthenium-carbene complex:

bis(tricyclohexylphosphine)benzylidene ruthenium dichloride.

A 2 mm thick high density polyethylene sheet was prepared, and after surface degreasing, was cut into two pieces 25 mm X 100 mm. The end of one of these cut pieces (area: 12.5 mm X 25 mm) was painted with an adhesive composition blended so that the mol ratio of the bis(tricyclohexylphosphine)benzylidene ruthenium dichloride was 1/10000 in respect to the polymerizable monomer, and then the other piece had its end (area: 12.5 X 25 mm) laid over and pasted on, and the product was given heat treatment at 80°C for 5 minutes under application of 490 kPa pressure to make a test piece. The adhesion work time including heat treatment was 7 minutes.

[0043] <Comparative Example 3> A 2 mm thick high density polyethylene sheet was prepared, and after surface degreasing, two 25 mm X 100 mm cut pieces were made, the ends of the two cut pieces (areas: 12.5 mm X 25 mm) were dipped in a primer comprising a 5% acetone solution of 2,4-lutidine and then withdrawn, and were let stand for a while to volatize the acetone. Then, ethylcyanoacrylate was painted onto the primer treated surface (area: 12.5 mm X 25 mm) of one cut piece, after which the primer treated surface (area: 12.5 mm X 25 mm) of the other cut piece was laid over and pasted, and the combined pieces were let stand for 5 hours under application of 490 kPa pressure to make a test piece. The adhesion work time extended to more than 5 hours.

[0044] <Comparative Example 4> A 2 mm thick high density polyethylene sheet was prepared, and after surface degreasing, two 25 mm X 100 mm cut pieces were made, the ends of the two cut pieces (areas: 12.5 mm X 25 mm) were dipped in a primer comprising a 5% acetone solution of 3,4-lutidine and then withdrawn, and were let stand for a while to volatize the acetone. Then, ethylcyanoacrylate was painted onto the primer treated surface (area: 12.5 mm X 25 mm) of one cut piece, after which the primer treated surface (area: 12.5 mm X 25 mm) of the other cut piece was laid over and pasted, and the combined pieces were let stand for 5 hours under application of 490 kPa pressure to make a test piece. The adhesion work time extended to more than 5 hours.

[0045] <Example 6>

Polymerizable monomer: 90 mol% dicyclopentadiene (90.8 wt%), 10 mol% 5-ethylidene-2-norbornene (9.2 wt%),
Ruthenium vinylidene complex: complex of the aforesaid formula III.
A 3 mm thick polydicyclopentadiene sheet was prepared, and after surface degreasing, was cut into two pieces 25 mm X 100 mm. The end of one of these cut pieces (area: 12.5 mm X 25 mm) was painted with an adhesive composition blended so that the mol ratio of the ruthenium vinylidene complex was 1/5000 in respect to the polymerizable monomer, and then the other piece had its end (area: 12.5 X 25 mm) laid over and pasted on, and the product was given heat treatment at 80°C for 15 minutes under application of 490 kPa

pressure to make a test piece. The adhesion work time including heat treatment was 17 minutes.

[0046] <Comparative Example 5> A 3 mm thick polydicyclopentadiene sheet was prepared, and after surface degreasing, two 25 mm X 100 mm cut pieces were made, the ends of the two cut pieces (areas: 12.5 mm X 25 mm) were dipped in a primer comprising a 5% acetone solution of 2,4-lutidine and then withdrawn, and were let stand for a while to volatize the acetone. Then, ethylcyanoacrylate was painted onto the primer treated surface (area: 12.5 mm X 25 mm) of one cut piece, after which the primer treated surface (area: 12.5 mm X 25 mm) of the other cut piece was laid over and pasted, and the combined pieces were let stand for 5 hours under application of 490 kPa pressure to make a test piece. The adhesion work time extended to more than 5 hours.

[0047] <Comparative Example 6> A 3 mm thick polydicyclopentadiene sheet was prepared, and after surface degreasing, two 25 mm X 100 mm cut pieces were made, the ends of the two cut pieces (areas: 12.5 mm X 25 mm) were dipped in a primer comprising a 5% acetone solution of 3,4-lutidine and then withdrawn, and were let stand for a while to volatize the acetone. Then, ethylcyanoacrylate was painted onto the primer treated surface (area: 12.5 mm X 25 mm) of one cut piece, after which the primer treated surface (area: 12.5 mm X 25 mm) of the other cut piece was laid over and pasted, and the combined pieces were let

stand for 5 hours under application of 490 kPa pressure to make a test piece. The adhesion work time extended to more than 5 hours. [0048] The tensile shear adhesive strengths of the test pieces obtained in the above Examples 1 to 6 and Comparative Examples 1 to 6 were measured (based on JIS K 6850), and the results are shown in Table 1 to Table 3 below.

[0049]

[Table 1]

	Ex. 1	Ex. 2	Comp. Ex. 1	Comp. Ex. 2
Average values (n = 5)	561.9	569.8	227.5	246.1
Maximum values	588.4	600.1	324.6	300.0
Minimum values	524.7	517.8	152.0	183.4
Material adhered	Polypropylene			

Numerical values in the table show tensile shear adhesive strength (N/cm²)

[0050]

[Table 2]

	Ex. 3	Ex. 4	Ex. 5	Comp. Ex. 3	Comp. Ex. 4
Average values (n = 5)	552.1	545.2	532.5	208.9	204.0
Maximum values	573.7	560.9	553.1	290.3	281.5
Minimum values	512.9	518.8	506.0	150.0	143.2
Material adhered	High density polyethylene				

Numerical values in the table show tensile shear adhesive strength (N/cm²)

[0051]

[Table 3]

	Ex. 6	Comp. Ex. 5	Comp. Ex. 6
Average values (n = 5)	937.5	135.3	127.5
Maximum values	963.0	151.0	153.0
Minimum values	914.0	105.9	108.9
Material adhered	Polydicyclopentadiene		

Numerical values in the table show tensile shear adhesive strength (N/cm²)

[0052] As is apparent from the results above, it will be understood that the adhesive for olefin resins of the present invention is capable of displaying good adhesiveness on PP and HDPE even when treatment with primer is not done, and it has superior workability in having a short time until display of adhesive strength in addition to not requiring primer treatment.

[0053]

[Effect Of The Invention] As explained above, following the present invention makes it possible in short time periods to strongly adhere sets of olefin resins that priorily had problems in adhesiveness.

[0054] Consequently when the adhesive of the present invention is used, it is possible to firmly adhere olefin resin products that formerly could only be given a strong joining force by mechanical means such as screws. As a result, it becomes possible to expand the field of utilization of olefin resin products. Also, since it is a

solventless reaction type adhesive, it reduces burdens on properties and the environment compared to solvent type adhesives.

End.